

Le A 33 726-US

- 1 -

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IN THE UNITED STATES PATENT OFFICE

APPLICANT: JOSEF-WALTER STAWITZ ET AL.  
SERIAL NO.: 10/009,749  
FILED: May 12, 2001  
TITLE: Use of Cu-Phthalocyanine Sulfonamides as a dye for  
write-once optical data storage means

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DECLARATION B

I, JOSEF W. Stawitz, declare:

that I am a German citizen resident at Am Hagen, 51519 Odenthal, Germany;

that I am a chemist having graduated with a degree of Doctor rer. nat. from the University of Würzburg, Germany in 1978;

that I have since been concerned with the preparation of organic dyestuffs;

that I am one of the joint inventors of US Patent Application Serial No. 10/009 749 filed on May 12, 2001;

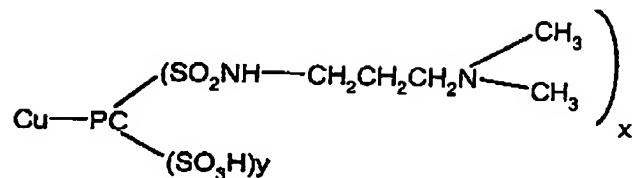
that I have read the Office Action of August 18, 2003 and the references cited therein;

that the following dyestuff mixtures were tested under my supervision.

Le A 33 726-US

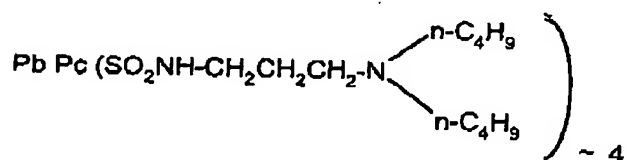
- 2 -

## I. Dye of the formula



- Ia)  $x \sim 3.7$ ;  $y \sim 0.3$  according to US-Patent Application Serial No. 10/009749, similar to Exp. 1 (present invention)
- Ib)  $x \sim 3.0$ ;  $y \sim 0$  according to US-Patent Application Serial No. 10/009749, Exp. 2 (present invention)
- Ic)  $x \sim 4.0$ ;  $y \sim 0$  according to US-Patent Application Serial No. 10/009749, covered by claim 1.

## IV. Dye of the formula



according to JP-A-63-307987 (dye mentioned below exp. 14).

The dye of the formula IV was intended to be synthesized by reaction of the Pb-phthalocyanine with chlorosulfuric acid at 100 to 130 °C. At a temperature range of 100 to 110 °C, although normally only small sulfonation occurs there, it already leads to the formation of PbSO<sub>4</sub> and metal-free phthalocyanine (which means a decomposition reaction took place. At higher temperatures (120 to 130° C) required for normal phthalocyanines with a substitution made of 3 to 4 the decomposition becomes stronger until finally

Le A 33 726-US

- 3 -

the decomposition to phthalic acid derivatives is complete so that the Pb-compound of JP-A-63-307987, could not be synthesized.

This observation was already made earlier by P. A. Barrett et al. (J. Chem. Soc. 1936, P. 1719-1736), in particular page 1725, line 7 – 10.

“On the other hand, longer or smaller covalent metals, such as manganese (1-18) and lead (1-75) are displaced by acids.”

Conclusively, as no method for the preparation of such a lead-phthalocyanine as mentioned in JP-'987 is disclosed therein, the compound seems to be not possible to be synthesized. This compound has to be considered as an accidentally disclosure in the JP-reference.

Le A 33 726-US

- 4 -

Tests

The following tests were made in order to have evidences whether or not applicability of the dye by using the spin coating technique is possible. For this, solution of the dyestuffs Ia, Ib, Ic and IV respectively in most used solvents for spin coating, were made.

Such obtained solutions were given to a filter paper to see the chromatographic behavior. The respective filter papers are attached to this declaration.

- X (present invention): cpd. Ia) (5% by weight) was dissolved in tetrafluoro-propanol. The product was completely dissolved.
- Y (present invention): cpd. Ib) (5% by weight) was dissolved in tetrafluoro-propanol. The product was completely dissolved.
- Z (present invention): cpd. Ic) (5% by weight) was dissolved in tetrafluoro-propanol. The product was completely dissolved.

Result

As can be seen from the attached card (respective filter papers are mounted thereon). The compounds Ia) to Ic) can be completely dissolved in the most common solvents used for spin coating respective of the different number of SO<sub>3</sub>H groups in the molecule.

In addition a substituted Pb-phthalocyanine can not be synthesized due to decomposition sensibility as also know from the prior art.

Le A 33 726-US

- 5 -

Conclusion

The unexpected difference of the dye Ia) and III in favour of cpd. Ia) as shown in the declaration of Dr. Stawitz dated Feb. 9, 2004, is exclusively based on the different center atom of the phthalocyanine used. Under the tested conditions no different behaviour is observed due to different numbers of SO<sub>3</sub>H groups in the molecule.

The undersigning declarant declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date:

Signature:

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Josef W. Stawitz



X  
(Ia)



Y  
(Ia)



Z  
(Ia)